IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of : MATSUO Shinji et al

Serial Number : 10/060,203

Filed : February 1, 2002

For : Organic Electroluminescent Material and Device Made Therefrom

Group Art Unit : 1774

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner

Washington, D.C. 20231

Sir:

Now comes MIYAZAKI Hiroshi who declares and says that:

1. I am an inventor of United States Patent Application Serial Number 10/060,203.

2. I graduated from Hokkaido University, Faculty of Science, Department of Chemistry in 1984, and finished the master's course thereof in 1986, and studied Chemistry of Organic Synthesis.

3. I have been employed by Nippon Steel Chemical Co., Ltd. since 1986, and investigated of Organic electroluminescent materials (OELM) from 1996.

4. I have conducted an extensive search for patent literature relating to the methods for preparing and purifying triarylamines useful for OELM while covering the data bases of the United States Patent Office, Japanese Patent Office, WPI and

CAS.

As a result, I have found the following documents and others relating to the methods for preparing triarylamines for use as OELM.

JP11288508 A2 19990824

JP10017531 A2 19980120

In addition, I have found the following documents relating to the methods for preparing triarylamines for potential use as OELM.

EP802173 A1 19971022

USP5648542 A 19970715

EP34425 A2 19810826

USP4764625 A 19880816

5. The methods used in the aforementioned documents are based either on

a) the Ullmann reaction of an arylamine with the corresponding aryl halide effected under the basic condition in the presence of copper or a copper complex as a catalyst or

b) the coupling reaction of an arylamine with the corresponding aryl halide effected under the basic condition in the presence of a palladium complex containing a trialkylphosphine as a ligand.

The reaction a) is described in paragraph 【0009】 and Synthetic Example 1 of the specification of the present application while the reaction b) is described in paragraph 【0024】 and Synthetic Example 2 of the specification of the present application.

As any of these methods yields numerous byproducts upon completion of the reaction, the main product is purified by chromatographic separation or recrystallization or a combination of the two.

The typical byproducts are the ones referred to as Compounds (A) and (B) in the specification of the present application and this fact is shown in paragraphs 【0022】 and 【0024】 and Synthetic Examples 1 and 2 of the specification of the

present application.

6. I have carried out the following supplementary experiments.

Supplementary Experiments

Experiment 1

An experiment on purification was carried out as follows by recrystallizing the NPB obtained by chromatographic separation in Synthetic Example 1 of the specification of the present application.

In a 300-ml three-necked flask equipped with a thermometer and a condenser were placed 10 g of the column-treated NPB containing 4% of compound (A) and 7% of compound (B) and 200 g of DMF and the contents were heated with stirring until the DMF started to reflux (oil bath temperature, approximately 180 ℃). The heating was stopped after the dissolution of the contents was confirmed and then the contents were allowed to cool to room temperature (approximately 20 ℃) to separate crystals.

The crystals were filtered under reduced pressure by the use of a Kiriyama funnel and washed successively with approximately 50 g of DMF and approximately 100 g of methanol on the funnel. The crystals thus obtained were dried under reduced pressure to give 6.4 g of purified product.

The product (NPB) thus purified by recrystallization contained 0.7% of compound (A) and 3.5% of compound (B) when analyzed by HPLC.

Conclusions

The following conclusions can be drawn from the aforementioned experimental results.

1. The aforementioned search for patent literature indicates that the methods for preparing triarylamines are based on the Ullmann reaction or the coupling

reaction by the use of a palladium complex regardless of whether the triarylamines are intended for use as OELM or not.

2. The triarylamines prepared by these methods are purified by chromatographic separation or recrystallization or by a combination of the two as they contain a large amount of impurities.

3. However, purification by chromatographic separation and recrystallization does not sufficiently reduce the amounts of compound (A) and compound (B) as indicated by the aforementioned experiment.

This insufficient reduction of the byproducts seems to be due to the fact that compound (A) and compound (B) are triarylamines similar to the main component and there is a small difference between the main component and compound (A) or compound (B) in the adsorbability in chromatographic separation and also in the solubility in a solvent to be used in recrystallization.

4. Therefore, it is likely that the triarylamines regarded practical as OELM at the time of filing of the present application contained 1% or more (or 0.5% or more) of compound (A) and 2% or more (or 1% or more) of compound (B).

5. On the other hand, the method adopted in the refining example of the present invention is based on sublimation/distillation. According to this method, crude triarylamines are purified by sublimation or evaporation under heat followed by solidification in the collecting zone where the temperature is set at the solidification temperature of the target triarylamines. This method is capable of separating the impurities (A) and (B) efficiently without degenerating the triarylamines and purifying the triarylamines to a quality suitable for use as OELM. The advantage of this purifying method is shown by Purifying Examples 1 and 2 of the specification of the present application.

I, the undersigned petitioner, further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

25th day of March 2004.

H. Miyazaki

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